

III. REVIEW OF BASIC QUANTUM MECHANICS: TWO-LEVEL QUANTUM SYSTEMS:

The literature of quantum optics and laser spectroscopy abounds with discussions of the two-level (two-state) system. This emphasis comes about because the interaction of such systems with the electromagnetic field may be treated in great detail to obtain valuable analytic results and, hopefully, the analysis of two-level systems generates insights that may be extended to more realistic situations. Fortunately, there are several important instances in which the application of the two-level model provides a very good approximation to a more complete theory. In the following, we label the upper level of the system by the letter **a** and the lower by the letter **b**. From Equation [I-12a] we write, specifically, the wave function of the two level system as

$$(\vec{r}, t) = \langle \vec{r} | (t) \rangle = C_a(t) u_a(\vec{r}) \exp(-i E_a t) + C_b(t) u_b(\vec{r}) \exp(-i E_b t) \quad [\text{III-1}]$$

where we know from Equation [I-12b] that the time varying coefficients satisfy, in general, the following equations:

$$\dot{C}_a(t) = -\frac{i}{\hbar} \{ C_a(t) \langle E_a | \mathcal{H}_1(t) | E_a \rangle + C_b(t) \langle E_a | \mathcal{H}_1(t) | E_b \rangle \exp(i E_{ab} t) \} \quad [\text{III-2a}]$$

$$\dot{C}_b(t) = -\frac{i}{\hbar} \{ C_b(t) \langle E_b | \mathcal{H}_1(t) | E_b \rangle + C_a(t) \langle E_b | \mathcal{H}_1(t) | E_a \rangle \exp(i E_{ba} t) \} \quad [\text{III-2b}]$$

If we take the interaction to be the electric dipole interaction with an applied electric field we may write

$$\mathcal{H}_1(t) = -\vec{\mathbf{p}}_s \cdot \vec{\mathbf{E}}(\vec{\mathbf{R}}_s, t) = -e \vec{\mathbf{r}} \cdot \vec{\mathbf{E}}(\vec{\mathbf{R}}_s, t) \quad [\text{III-3a}]$$

where $\vec{\mathbf{R}}_s$ denotes the position of the center of the two-level system or atom.¹⁴ Thus we write

$$\langle E_i | \mathcal{H}_1(t) | E_j \rangle = \mathcal{V}_{ij} = -\langle E_i | e \vec{\mathbf{r}} | E_j \rangle \cdot \vec{\mathbf{E}}(\vec{\mathbf{R}}_s, t) \quad [\text{III-3b}]$$

In all but the most bizarre circumstances we may use persuasive symmetry arguments to reason that

$$\langle E_i | e \vec{\mathbf{r}} | E_i \rangle = 0$$

Thus Equations [III-2] reduce to

$$\dot{C}_a(t) = -\frac{i}{\hbar} C_b(t) \mathcal{V}_{ab} \exp(i \omega_{ab} t) \quad [\text{III-4a}]$$

$$\dot{C}_b(t) = -\frac{i}{\hbar} C_a(t) \mathcal{V}_{ab} \exp(-i \omega_{ab} t) \quad [\text{III-4b}]$$

where $\mathcal{V}_{ab} = -\langle E_a | e \vec{\mathbf{r}} | E_b \rangle \cdot \vec{\mathbf{E}}(\vec{\mathbf{R}}_s, t) = -\langle E_b | e \vec{\mathbf{r}} | E_a \rangle \cdot \vec{\mathbf{E}}(\vec{\mathbf{R}}_s, t)$.

RABI FLOPPING -- WITHOUT DAMPING:

For an oscillatory applied field

$$\mathcal{V}_{ab} = -\langle E_a | e \vec{\mathbf{r}} | E_b \rangle \cdot \vec{\mathbf{E}}_0 \cos \omega_r t = -\frac{1}{2} \langle E_a | e \vec{\mathbf{r}} | E_b \rangle \cdot \vec{\mathbf{E}}_0 \exp(-i \omega_r t) + c.c. \quad [\text{III-5}]$$

we see, in the *rotating-wave approximation*, that

$$\begin{aligned} \dot{C}_a(t) &= \frac{i}{2\hbar} \langle E_a | e \vec{\mathbf{r}} | E_b \rangle \cdot \vec{\mathbf{E}}_0 C_b(t) \exp[i(\omega_{ab} - \omega_r)t] \\ &= \frac{i}{2} \langle E_a | e \vec{\mathbf{r}} | E_b \rangle \cdot \vec{\mathbf{E}}_0 C_b(t) \exp[i(\omega_{ab} - \omega_r)t] \end{aligned} \quad [\text{III-6a}]$$

¹⁴ The use of this form of interaction needs considerable elaboration, but we defer that discussion until later.

$$\begin{aligned}\dot{C}_b(t) &= \frac{i}{2\hbar} E_o C_a(t) \exp[-i(\omega_{ab} - \omega_r)t] \\ &= \frac{i}{2} \omega_o C_a(t) \exp[-i(\omega_{ab} - \omega_r)t]\end{aligned}\quad [\text{III-6b}]$$

where $\omega_o = E_o/\hbar$ defines the so called **Rabi flopping frequency** which is, of course, a measure of the strength of the electromagnetic interaction. Clearly, the coupling terms have maximum effect when the frequency of the applied field is resonant with the level splitting. In most treatments the *frequency detuning* of the field is expressed as $\omega_{ab} - \omega_r$ and the system's wave function - *i.e.* Equation [III-1] - is written in terms of slightly modified time varying coefficients by transforming to the *rotating frame of reference* - *viz.*

$$C_a(t) = \tilde{C}_a(t) \exp\left(i \frac{1}{2} \omega_o t\right) \quad [\text{III-7a}]$$

$$C_b(t) = \tilde{C}_b(t) \exp\left(-i \frac{1}{2} \omega_o t\right) \quad [\text{III-7b}]$$

$$(\vec{r}, t) = \tilde{C}_a(t) \exp\left[i \left(\frac{1}{2} \omega_o - \omega_a\right) t\right] u_a(\vec{r}) + \tilde{C}_b(t) \exp\left[i \left(-\frac{1}{2} \omega_o - \omega_b\right) t\right] u_b(\vec{r}) \quad [\text{III-7c}]$$

The coupling terms in this rotating frame of reference become

$$\dot{\tilde{C}}_a(t) = \frac{1}{2} i \left\{ -\tilde{C}_a(t) + \omega_o \tilde{C}_b(t) \right\} \quad [\text{III-8a}]$$

$$\dot{\tilde{C}}_b(t) = \frac{1}{2} i \left\{ \tilde{C}_b(t) + \omega_o \tilde{C}_a(t) \right\} \quad [\text{III-8b}]$$

or in matrix form

$$\frac{d}{dt} \begin{pmatrix} \tilde{C}_a(t) \\ \tilde{C}_b(t) \end{pmatrix} = \frac{i}{2} \begin{pmatrix} - & \omega_o \\ \omega_o & \end{pmatrix} \begin{pmatrix} \tilde{C}_a(t) \\ \tilde{C}_b(t) \end{pmatrix} = \frac{i}{2} \mathbf{M} \tilde{\mathbf{C}}(t) . \quad [\text{III-8c}]$$

We look for a solution in the form $\tilde{\mathbf{C}}(t) = \tilde{\mathbf{C}}(0) \exp\left(\frac{1}{2} i \mathbf{R} t\right)$ where \mathbf{R} is the generalization of the Rabi flopping frequency. Therefore, the condition

$$\det[\mathbf{M} - \mathbf{R} \mathbf{I}] = 0$$

yields the *generalized Rabi flopping frequency*

$$\mathbf{R} = \sqrt{\omega_o^2 + \left| \frac{\mathbf{R}_o}{\omega_o} \right|^2} \quad [\text{III-9a}]$$

and the general **non-damped** time evolving wave function

$$\begin{pmatrix} \tilde{C}_a(t) \\ \tilde{C}_b(t) \end{pmatrix} = \begin{pmatrix} \cos\left(\frac{1}{2} \mathbf{R} t\right) - \frac{i}{\mathbf{R}} \sin\left(\frac{1}{2} \mathbf{R} t\right) & \frac{i}{\mathbf{R}_o} \sin\left(\frac{1}{2} \mathbf{R} t\right) \\ \frac{i}{\mathbf{R}_o} \sin\left(\frac{1}{2} \mathbf{R} t\right) & \cos\left(\frac{1}{2} \mathbf{R} t\right) + \frac{i}{\mathbf{R}} \sin\left(\frac{1}{2} \mathbf{R} t\right) \end{pmatrix} \begin{pmatrix} \tilde{C}_a(0) \\ \tilde{C}_b(0) \end{pmatrix} \quad [\text{III-9b}]$$

RABI FLOPPING -- WITH DAMPING:

Neglected interactions (e.g. spontaneous emission, collisions, thermal fluctuations) limit lifetime of a state of a two-level system. One class of lifetime limiting interactions may be described **phenomenologically** by adding decay terms to the equations of motion - *i.e.* to Equations [III-8] - as follows:

$$\dot{\tilde{C}}_a(t) = -\frac{1}{2} \begin{pmatrix} \omega_a + i & \mathbf{R}_o \\ \mathbf{R}_o & \omega_b \end{pmatrix} \begin{pmatrix} \tilde{C}_a(t) \\ \tilde{C}_b(t) \end{pmatrix} \quad [\text{III-10a}]$$

$$\dot{\tilde{C}}_b(t) = -\frac{1}{2} \begin{pmatrix} \omega_b - i & \mathbf{R}_o \\ \mathbf{R}_o & \omega_a \end{pmatrix} \begin{pmatrix} \tilde{C}_a(t) \\ \tilde{C}_b(t) \end{pmatrix} \quad [\text{III-10b}]$$

or

$$\frac{d}{dt} \tilde{\mathbf{C}}(t) = \frac{d}{dt} \begin{pmatrix} \tilde{C}_a(t) \\ \tilde{C}_b(t) \end{pmatrix} = \frac{i}{2} \begin{pmatrix} -\omega_a & \mathbf{R}_o \\ \mathbf{R}_o & \omega_b \end{pmatrix} \begin{pmatrix} \tilde{C}_a(t) \\ \tilde{C}_b(t) \end{pmatrix} = \frac{i}{2} \mathbf{M} \tilde{\mathbf{C}}(t) \quad [\text{III-10c}]$$

Again we look for a solution in the form $\tilde{\mathbf{C}}(t) = \tilde{\mathbf{C}}(0) \exp\left(\frac{1}{2}i\omega t\right)$ so that

$$\det[\mathbf{M} - \mathbf{I}] = 0$$

$$\text{or} \quad \left[\frac{1}{2}(\omega_a + \omega_b) - \left(-\frac{1}{2}i(\omega_a - \omega_b) \right) + \left| \frac{\mathbf{R}}{\omega} \right|^2 \right] = 0$$

$$\text{Therefore} \quad \omega = \frac{1}{2}(\omega_a + \omega_b) \pm \sqrt{\left[-\frac{1}{2}i(\omega_a - \omega_b) \right]^2 + \left| \frac{\mathbf{R}}{\omega} \right|^2} \quad [\text{III-11a}]$$

-- where the we refer to $\omega_{ab} = \frac{1}{2}(\omega_a + \omega_b)$ as the **average decay rate constant** and $\omega_{\mathbf{R}} = \sqrt{\left[-\frac{1}{2}i(\omega_a - \omega_b) \right]^2 + \left| \frac{\mathbf{R}}{\omega} \right|^2}$ as the **generalized complex Rabi flopping frequency** -- and the general time evolving wave function -- in the rotation frame -- may be written

$$\begin{pmatrix} \tilde{C}_a(t) \\ \tilde{C}_b(t) \end{pmatrix} = \begin{pmatrix} \cos\left(\frac{1}{2}\omega_{\mathbf{R}} t\right) - \frac{\left[\frac{1}{2}(\omega_a - \omega_b) + i\right]}{\omega_{\mathbf{R}}} \sin\left(\frac{1}{2}\omega_{\mathbf{R}} t\right) \\ \frac{i}{\omega_{\mathbf{R}}} \sin\left(\frac{1}{2}\omega_{\mathbf{R}} t\right) \end{pmatrix} \exp\left(-\frac{1}{2}\omega_{ab} t\right) \begin{pmatrix} \tilde{C}_a(0) \\ \tilde{C}_b(0) \end{pmatrix} \quad [\text{III-11b}]$$

DENSITY MATRIX TREATMENT OF A TWO-LEVEL SYSTEMS:

Recall from Equation [II-30]

$$\dot{\rho} = \frac{i}{\hbar} [\rho, \mathcal{H}] = \frac{i}{\hbar} \{ \mathcal{H} - \mathcal{H} \}$$

Using $|a\rangle\langle a| + |b\rangle\langle b| = 1$ we may write

$$\dot{\rho} = \frac{i}{\hbar} \left\{ \left(|a\rangle\langle a| + |b\rangle\langle b| \right) \mathcal{H} - \mathcal{H} \left(|a\rangle\langle a| + |b\rangle\langle b| \right) \right\} \quad [\text{III-12a}]$$

If $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1$, then

$$\dot{\cdot} = \frac{i}{\hbar} \left\{ \begin{aligned} & [E_a |a\rangle\langle a| + E_b |b\rangle\langle b| + |a\rangle\langle a| \mathcal{H}_1 + |b\rangle\langle b| \mathcal{H}_1] \\ & - [E_a |a\rangle\langle a| + E_b |b\rangle\langle b| + \mathcal{H}_1 |a\rangle\langle a| + \mathcal{H}_1 |b\rangle\langle b|] \end{aligned} \right\} \quad [\text{III-12b}]$$

Writing matrix elements -- *i.e.* representatives -- of the density operator

$$\langle a | \dot{\cdot} | a \rangle = \frac{i}{\hbar} \{ \langle a | |b\rangle\langle b| \mathcal{H}_1 | a \rangle - \langle a | \mathcal{H}_1 | b \rangle \langle b | |a \rangle \} \quad [\text{III-13a}]$$

$$\langle b | \dot{\cdot} | b \rangle = \frac{i}{\hbar} \{ \langle b | |a\rangle\langle a| \mathcal{H}_1 | b \rangle - \langle b | \mathcal{H}_1 | a \rangle \langle a | |b \rangle \} \quad [\text{III-13b}]$$

$$\begin{aligned} \langle a | \dot{\cdot} | b \rangle = \frac{i}{\hbar} \{ & -(E_a - E_b) \langle a | |b \rangle + [\langle a | |a \rangle - \langle b | |b \rangle] \langle a | \mathcal{H}_1 | b \rangle \\ & + \langle a | |b \rangle [\langle b | \mathcal{H}_1 | b \rangle - \langle a | \mathcal{H}_1 | a \rangle] \} \end{aligned} \quad [\text{III-13c}]$$

We may again assume, by symmetry arguments, that $\langle a | \mathcal{H}_1 | a \rangle = \langle b | \mathcal{H}_1 | b \rangle = 0$ so that the equations of motion for the elements of the density reduce to

$$\dot{\cdot}_{bb} - \dot{\cdot}_{aa} = \left[i \hbar^{-1} 2 \mathcal{V}_{ab \quad ba} + c.c. \right] \quad [\text{III-14a}]$$

$$\dot{\cdot}_{ab} = -i \mathcal{V}_{ab \quad ab} + i \hbar^{-1} \mathcal{V}_{ab} \left[\dot{\cdot}_{aa} - \dot{\cdot}_{bb} \right] \quad [\text{III-14b}]$$

To introduce an element of reality, we add to these equations a pair of the most ***intuitively satisfying*** damping terms (a useful attribute of density matrix formulations) -- *viz.*

$$\dot{\cdot}_{bb} - \dot{\cdot}_{aa} = \left[i \hbar^{-1} 2 \mathcal{V}_{ab \quad ba} + c.c. \right] - \left[\left(\dot{\cdot}_{bb} - \dot{\cdot}_{aa} \right) - \left(\dot{\cdot}_{bb} - \dot{\cdot}_{aa} \right)_0 \right] \quad [\text{III-15a}]$$

$$\dot{a}_{ab} = -i \omega_{ab} a_{ab} + i \hbar^{-1} \mathcal{V}_{ab} \left[a_{aa} - a_{bb} \right] - \dot{a}_{ab} \quad [\text{III-15b}]$$

Transform these equations to a **rotating frame** by taking $a_{ab} = \tilde{a}_{ab} \exp(-i \omega_r t)$ where \tilde{a}_{ab} is assumed to be a slowly varying function of time which satisfies the equations of motion

$$\dot{\tilde{a}}_{ab} = -i \omega_{ab} \tilde{a}_{ab} + i \hbar^{-1} \mathcal{V}_{ab} \exp(i \omega_r t) \left[a_{aa} - a_{bb} \right] - \dot{\tilde{a}}_{ab} \quad [\text{III-16a}]$$

$$\begin{aligned} \dot{a}_{bb} - \dot{a}_{aa} &= \left[i \hbar^{-1} 2 \mathcal{V}_{ab} \exp(i \omega_r t) \tilde{a}_{ba} + c.c. \right] \\ &\quad - \left[\left(a_{bb} - a_{aa} \right) - \left(a_{bb} - a_{aa} \right)_0 \right] \end{aligned} \quad [\text{III-16b}]$$

With $\mathcal{V}_{ab} = -\left[\chi_2 \hbar \omega_0 \exp(-i \omega_r t) + c.c. \right]$ and ignoring terms proportional to $\exp(\pm i 2 \omega_r t)$ -- **i.e. the rotating wave approximation** - we find that

$$\dot{\tilde{a}}_{ab} = -i \left(\omega_{ab} - \omega_r \right) \tilde{a}_{ab} - i \left(\chi_2 \hbar \omega_0 \right) \left[a_{aa} - a_{bb} \right] \quad [\text{III-17a}]$$

$$\dot{a}_{bb} - \dot{a}_{aa} = \left[-i \hbar \omega_0 \tilde{a}_{ba} + c.c. \right] - \left[\left(a_{bb} - a_{aa} \right) - \left(a_{bb} - a_{aa} \right)_0 \right] \quad [\text{III-17b}]$$

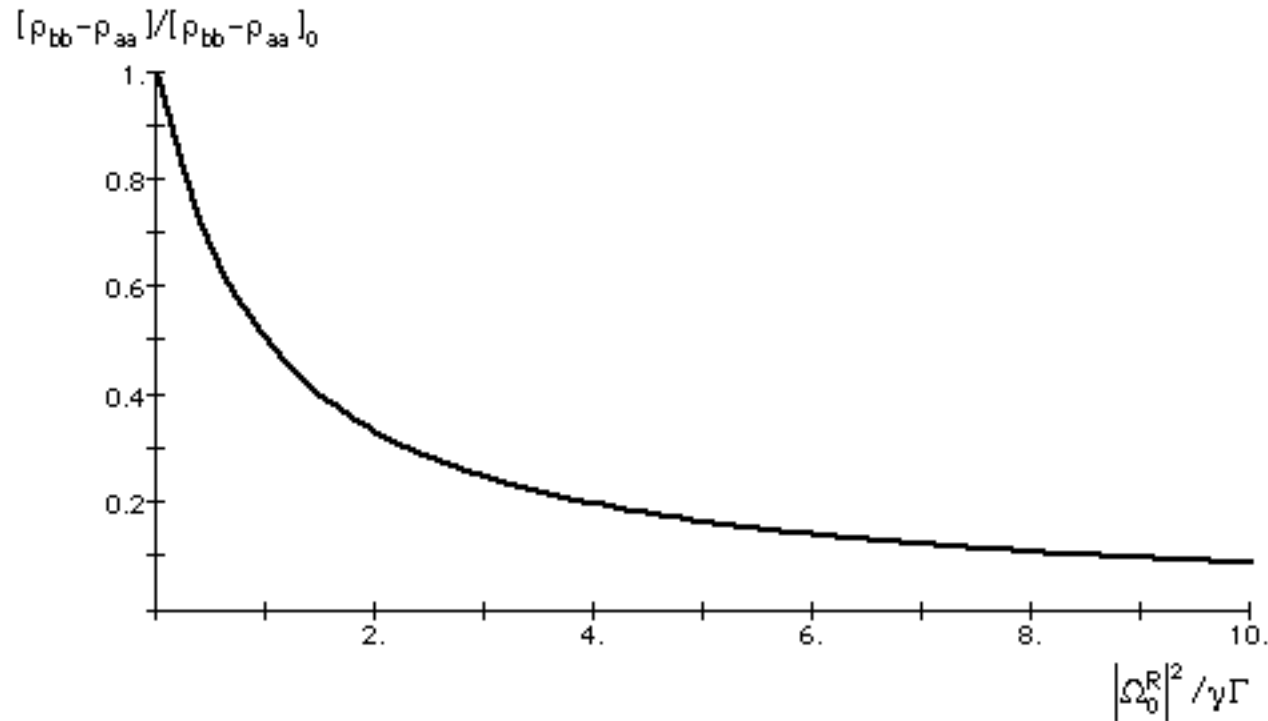
Steady state behavior: If we take all time derivatives in these equations equal to zero, we obtain

$$\tilde{a}_{ab} = \frac{i \hbar \omega_0 \left[a_{bb} - a_{aa} \right]}{2 \left(i \omega_{ab} - \omega_r \right)} = \frac{\hbar \omega_0 \left(\omega_{ab} + i \right) \left[a_{bb} - a_{aa} \right]}{2 \left(\omega_{ab}^2 + \omega_r^2 \right)} \quad [\text{III-18a}]$$

$$\left(a_{bb} - a_{aa} \right) = \left(a_{bb} - a_{aa} \right)_0 \frac{\omega_{ab}^2 + \omega_r^2}{\omega_{ab}^2 + \omega_r^2 + 1 + \left| \hbar \omega_0 \right|^2} \quad [\text{III-18b}]$$

or

$$\tilde{\rho}_{ab} = \frac{\gamma_2 \frac{\mathbf{R}}{\omega} \left[\begin{array}{c} \\ \end{array} + i \right] \left(\rho_{bb} - \rho_{aa} \right)_0}{\omega^2 + \gamma^2 + 1 + \left| \frac{\mathbf{R}}{\omega} \right|^2} \quad [\text{III-18c}]$$



We have graphed Equation [III-18b] at resonance - *i.e.* $\omega = 0$ - and, from the following expression, we see how the oscillatory polarization is **saturated** at high electromagnetic powers. In general, the **macroscopic polarization** is then given by¹⁵

¹⁵ That is to say

$$\langle \mathbf{P} \rangle = \left(\mathbf{P} \right)_{aa} + \left(\mathbf{P} \right)_{bb} = \mathbf{P}_{aa} \quad aa + \mathbf{P}_{ab} \quad ba + \mathbf{P}_{ba} \quad ab + \mathbf{P}_{bb} \quad bb = \mathbf{P}_{ab} \quad ba + \mathbf{P}_{ba} \quad ab$$

since $\mathbf{P}_{aa} = \mathbf{P}_{bb} = 0$ by symmetry.

If we graph Equation [III-18b] at resonance - *i.e.* $\omega = 0$ - we see how the oscillatory polarization is **saturated** at high electromagnetic powers - *viz.*

$$P = N \langle \text{dipole moment of system} \rangle = N \left[\mu_{ba} + \text{c.c.} \right] \quad [\text{III-19a}]$$

$$P = \frac{N \mu_0^2 \left(\frac{\omega}{\omega_0} - i \right) \left[\mu_{bb} - \mu_{aa} \right]}{2 \left(\frac{\omega}{\omega_0}^2 + 1 \right)} \exp(i \omega_r t) + \text{c.c.} \quad [\text{III-19b}]$$

$$P = \frac{N |\mu|^2 \left[\frac{\omega}{\omega_0} - i \right] \left[\mu_{bb} - \mu_{aa} \right]_0}{2 \hbar \left(\frac{\omega}{\omega_0}^2 + 1 \right) \left[1 + \left| \frac{\mu}{\mu_0} \right|^2 \right]} E_0 \exp(i \omega_r t) + \text{c.c.} \quad [\text{III-19c}]$$

THE VECTOR MODEL OF THE DENSITY MATRIX:

There is a set of **arguments by analogy** which is exceedingly valuable in treating transient excitation problems in optics. The basis for the analogy lies in the fact that Equations [III-16] are identical in form to the **famous Bloch equations of magnetic resonance**.¹⁶ If we make a transformation to a rotating frame -- *i.e.*, $M_{\pm} = \tilde{M}_{\pm} \exp(\mp i \omega_r t)$ -- the Bloch equations take on the form

¹⁶ Recall from the theory of magnetic resonance

$$d\vec{M}/dt = \gamma_{\text{mag}} \vec{M} \times \vec{H}$$

where γ_{mag} is the **gyromagnetic ratio**. This equation of motion is greatly simplified if it is written in terms of the circular polarizations $M_{\pm} = M_x \pm i M_y$ and $H_{\pm} = H_x \pm i H_y$ -- *viz.*

$$\dot{M}_{\pm} = \mp i \gamma_{\text{mag}} M_{\pm} H_z \pm i \gamma_{\text{mag}} M_z H_{\pm}$$

and

$$\dot{M}_z = i \frac{\gamma_{\text{mag}}}{2} [M_+ H_- - M_- H_+]$$

To these Bloch added the phenomenological longitudinal (thermal) relaxation time T_1 and transverse

$$\dot{\tilde{M}}_{\pm} = \mp i \tilde{M}_{\pm} \pm i \omega_{\text{mag}} \tilde{M}_z \exp(\pm i \omega_r t) H_{\pm} - \tilde{M}_{\pm}/T_2 \quad [\text{III-20a}]$$

$$\dot{M}_z = -i \frac{\omega_{\text{mag}}}{2} H_- \exp(-i \omega_r t) M_+ + c.c. - (M_z - M_{z0})/T_1 \quad [\text{III-20b}]$$

where $\omega_r = \omega - \omega_{\text{mag}}$. By comparing these Bloch equations with Equations [III-16] and [III-17] we see that we have identical problems if we make the identifications

$$\tilde{M}_+ \rightarrow \tilde{\rho}_{ab} \quad [\text{III-21a}]$$

$$\tilde{M}_z \rightarrow \tilde{\rho}_{bb} - \tilde{\rho}_{aa} \quad [\text{III-21b}]$$

$$\omega_{\text{mag}} H_z \rightarrow \omega_{ab} \quad [\text{III-21c}]$$

$$\omega_{\text{mag}} H_+ \rightarrow \hbar^{-1} \mathcal{V}_{ab} = -\gamma/2 \quad \mathbf{R}_0 \quad [\text{III-21d}]$$

$$1/T_1 = \quad [\text{III-21e}]$$

$$1/T_2 = \quad [\text{III-21f}]$$

In other words we have the equivalent equation of motion

$$\dot{\vec{M}} = \vec{M} \times \vec{\omega}_{\text{eff}} - \left(\vec{M} - \hat{z} M_z \right) - \hat{z} M_z \quad [\text{III-22}]$$

where the *effective Rabi precession field* $\vec{\omega}_{\text{eff}}$ is given by

$$\vec{\omega}_{\text{eff}} = \mathbf{R}_0 \hat{\mathbf{x}} - \hat{\mathbf{z}} \quad [\text{III-23}]$$

dephasing time T_2 so that

$$\dot{M}_{\pm} = \mp i \omega_{\text{mag}} M_{\pm} H_z \pm i \omega_{\text{mag}} M_z H_{\pm} - M_{\pm}/T_2$$

$$\dot{M}_z = i \frac{\omega_{\text{mag}}}{2} [M_+ H_- - M_- H_+] - (M_z - M_{z0})/T_1$$

